Nuclear magnetization and NMR in a ferromagnet when the internal magnetic field is cancelled by application of an external field

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The equilibrium state of nuclear magnetization and of NMR in a magnet is considered under conditions when the internal magnetic field at the nuclei $\mathbf{H}_{hf}$ is offset by an external field $\mathbf{H}$. For temperatures $T > T_c$ (T_c is the nuclear magnetic system ordering temperature), the nuclear magnetization $\boldsymbol{\mu}$ is collinear with the magnetic field $\mathbf{H}$ and its variation is given by the Brillouin formula. For $T < T_c$ a noncollinear situation is produced in the region $H \sim H_{hf}$, viz., the vector $\boldsymbol{\mu}$ rotates and its modulus is independent of H and is determined only by the electron-nuclear interaction constant and by the temperature. If $H = H_{hf}$, spontaneous nuclear magnetization sets in at the point $T = T_c$ as the temperature is lowered. It is shown that the critical temperature can be raised in anisotropic magnets. The features of excitation and recording of NMR at $H \sim H_{hf}$ and $T < T_c$ are investigated. Similar effects can appear also in a two-sublattice magnet. It is suggested that cancellation of the internal magnetic fields in a substance can be used in a search for arion interactions.

It is known that the internal magnetic fields at the nuclei of ferromagnetic media reach values 10^4–10^6 Oe.[1] In many materials the field at the nuclei is described by the simple formula $\mathbf{H}_{hf} = -A\,\mathbf{M}$, where A is the hyperfine interaction constant ($A > 0$) and $\mathbf{M}$ is the electron magnetization. The nuclear magnetization $\boldsymbol{\mu}$ is therefore antiparallel to the electron magnetization $\mathbf{M}$ in the equilibrium state. Clearly, application of an external magnetic field $H > H_{hf}$ can alter the equilibrium orientation of $\boldsymbol{\mu}$. To our knowledge, no experiments have been performed so far, although in modern strong-magnetic-field technology no difficulties are encountered in principle.

Our main purpose was a theoretical investigation of the effects occurring in the region where the internal magnetic field in the nuclei is cancelled out.

1. EQUILIBRIUM STATE OF AN ELECTRON-NUCLEAR MAGNETIC SYSTEM

To analyze the equilibrium state of a system in the $H \sim H_{hf}$ region, we write down the expression for the macroscopic energy density of an isotropic ferromagnet:

$$F = -\mathbf{H}(\mathbf{M}+\boldsymbol{\mu}) + A\mathbf{M}\boldsymbol{\mu}. \tag{1}$$

The electron and nuclear magnetizations acted upon respectively by the respective effective magnetic fields $\mathbf{H}_e$ and $\mathbf{H}_n$:

$$\mathbf{H}_e = -\partial F/\partial \mathbf{M} = \mathbf{H} - A\boldsymbol{\mu}, \quad \mathbf{H}_n = -\partial F/\partial \boldsymbol{\mu} = \mathbf{H} - A\mathbf{M}. \tag{2}$$

In the equilibrium state we have

$$[\mathbf{M}\mathbf{H}_e] = 0, \quad [\boldsymbol{\mu}\mathbf{H}_n] = 0. \tag{3}$$

We consider temperatures T that are low compared with the temperature for magnetic ordering of the electron system. The dependence of M on T and H can then be neglected, i.e., M is constant in Eqs. (3). We neglect next also the interaction between the nuclear spins; $\mu(H,T)$ is given then by the Brillouin function

$$\mu = \mu(H,\ T) = \mu_0 B_I(\mu_I H_n/kT). \tag{4}$$

Here I is the nuclear spin, $\mu I = \gamma\hbar I$ its magnetic moment, γ the gyromagnetic ratio, $\mu_0 = N_{\mu_I}$ the nuclear magnetization at $T = 0$, and N the nuclear spin density.

Let the external magnetic field $\mathbf{H}$ be directed along the z axis (Fig. 1). (Note that in equilibrium $\mathbf{H}$, $\mathbf{M}$, and $\boldsymbol{\mu}$ lie in the same plane, since $\mathbf{H}_n$ is in the plane of the vectors $\mathbf{H}$ and $\mathbf{M}$ and the nuclear magnetization is directed along $\mathbf{H}_n$.) We denote by θ and φ the polar angles of $\mathbf{M}$ and $\boldsymbol{\mu}$.

The coupled system of equations (3) and (4) has besides the simple collinear solution ($\theta = 0$, $\varphi = 0, \pi$), which we number 1, the noncollinear solution 2:

$$\sin\theta = \frac{\mu}{M}\sin\varphi, \quad \cos\varphi = \frac{(A\mu)^2 + H^2 - (AM)^2}{2HA\mu}, \qquad \mu = \mu_0 B_I(\mu_I A\mu/kT). \tag{5}$$

It can be seen that the expression for the modulus of the external magnetization has the same structure as the equation for the spontaneous magnetization of a Heisenberg ferromagnet in the mean-field approximation.[2] This equation is known to have a nontrivial solution at temperatures $T < T_c$, where

$$T_c = AN\gamma^2\hbar^2 I(I+1)/3k; \tag{6}$$

the plot of $\mu(T)$ is also well known.[2] It can be seen from the expression for $\cos\varphi$ that the second solution exists under the condition

$$A\mu(T) > |H_\Delta|, \quad H_\Delta = H - AM. \tag{7}$$

Note that μ is independent of H. It is easily verified that if $T < T_c$ and if inequality (7) holds, the solution 2 corresponds to a lower energy than solution 1.

We analyze now the change of the nuclear magnetiza-

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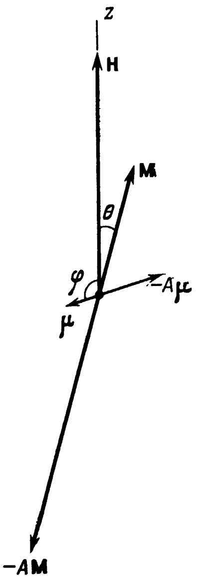

FIG. 1. Geometry of problem.

tion μ with increase of the external magnetic field H in the region $H \sim AM$. If $T > T_c$, the simple solution 1 is realized. If $H < AM$, the vector $\boldsymbol{\mu}$ is antiparallel to the z axis. As H increases, the value of μ decreases in accordance with the Brillouin formula, and at $H = AM$ we have $\mu = 0$. With further decrease of H the vector $\boldsymbol{\mu}$ becomes parallel to the z axis and its length increases.

The situation is substantially changed for $T < T_c$. So long as $H_\Delta \leqslant -A\mu$ the solution 1 considered above is realized as before. When H increases further, solution 2 is realized in the region $|H_\Delta| < A\mu$. This means that μ remains unchanged, but the vector $\boldsymbol{\mu}$ is turned. The angle φ decreases from π to 0 in accordance with Eq. (5). With further increase in H, the solution 1 is again realized in the region $H_\Delta \geqslant A\mu$.

2. SPONTANEOUS NUCLEAR MAGNETIZATION

We consider now the temperature dependence of the nuclear magnetization at constant H. It follows from the condition (7) that if $H_\Delta \geqslant A\mu_0$ we have solution 1 at all temperatures. In this case the vector $\boldsymbol{\mu}$ does not change direction when the temperature decreases, and its length increases in accordance with the Brillouin law. If $|H_\Delta| = H'_\Delta < A\mu_0$, there is always a value of T'_c (see Fig. 2) such that for $T < T'_c$ the condition (7) is met, i.e., the solution 2 is realized.

The most interesting situation is at $H_\Delta = 0$. In this case T'_c assumes its maximum value $T'_c = T_c$. At $T < T_c$ spontaneous nuclear magnetization sets in and is oriented along the x axis ($\varphi = \pi/2$). As the temperature is lowered the angle φ decreases slightly ($\cos\varphi \to \mu_0/2M$ as $T \to 0$).

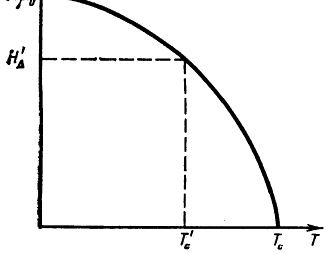

FIG. 2. The $|H_\Delta| = A\mu(T)$ curve bounds the region in which solution 2 is realized. The temperature T'_c corresponds to the fixed value $|H_\Delta| = H'_\Delta$.

Questions involving the onset of spontaneous nuclear magnetization in various media are extensively investigated in the literature at present.[3] The nuclear magnetic ordering due to direct or indirect interaction between nuclear spins is observed experimentally at ultralow temperatures $T \ll 1$ K. In our case, spontaneous nuclear magnetization sets in at $H_\Delta = 0$ because of indirect interaction of the nuclear spin via the electron ferromagnetic system. The critical temperature T_c is, naturally, very low. For cobalt, e.g., assuming $A \approx 160$, $N \approx 9\cdot 10^{22}$ cm^{-3}, $\gamma/2\pi \approx 10^3$ Hz/Oe, and $I = 7/2$ we get $T_c \approx 2\cdot 10^{-5}$ K.

Let us discuss the possibility of raising the critical temperature, obviously, the low value of T_c is due to the low susceptibility of the electron magnetic system in fields $H \sim AM$. Consider now a rare but apparently realistic situation, wherein the crystallographic-anisotropy field H_k is close in magnitude to the internal magnetic field at the nucleus. Application of an external field $H > H_k$ along the difficult-magnetization axis then results in a high electron magnetic susceptibility (since the system is close to the instability point), and we can expect an increase of T_c. (A similar method is used to make the NMR and FMR frequencies equal in thin ferromagnetic films[4].)

Let us examine this situation in greater detail. We add to Eq. (1) a term that describes uniaxial anisotropy with the easy axis collinear with the x axis:

$$\Delta F = -\beta M_x^2/2, \tag{8}$$

and assume that β is independent of T in the range of temperatures T of interest to us. Repeating the calculation described in the preceding section, we have again at $H > H_k = \beta M$ the collinear solution 1 and the solution 2 that can now be written out explicitly only for small θ:

$$\theta = \frac{\mu H}{M(H-H_k)}\sin\varphi, \qquad \cos\varphi = \frac{H_\Delta(H-H_k)}{A\mu H},$$
$$\mu = \mu_0 B_I[\mu_I A\mu H/kT(H-H_k)]. \tag{9}$$

These approximate expressions are valid under the condition

$$H - H_k \gg H\mu/M \sim A\mu. \tag{10}$$

It follows from the second equation in (9) that the condition for the transition from solution 1 to solution 2 can now be written in the form

$$A\mu \geqslant |H_\Delta|(H-H_k)/H. \tag{11}$$

The expression for the critical temperature T_c differs now from (6) by a factor $H/(H-H_k)$. By decreasing the difference $AM - H_k$ we can obtain in principle very high values of H_c, limited only by fluctuation processes.

Thus it seems that in media where H_k is close to AM the nuclear ferromagnetism can be "taken out" of the infralowtemperature region. It is possible that this situation will become realizable, for example, in compounds of rare-earth and $3d$ metals, where the anisotropy field is, according to published data,[5] of the order of 10^5 Oe (Ref. 5).

3. NUCLEAR MAGNETIC RESONANCE

We consider small oscillations of the nuclear magnetization about the equilibrium condition when a weak alternating magnetic field $\mathbf{h}=\mathbf{h}(t)$ is applied; we add a term $-\mathbf{h}(\mathbf{M}+\boldsymbol{\mu})$ to the expression for F. The NMR frequency is assumed low compared with that of the ferromagnetic resonance. The motion of the electron magnetization $\mathbf{M}$ is then described by the first expression (3), i.e., $\mathbf{M}$ executes quasi-static oscillations and follows the effective field $\mathbf{H}_c$. We write the nuclear-magnetization equation of motion in the form

$$\dot{\boldsymbol{\mu}}=-\gamma[\boldsymbol{\mu},\ \partial F/\partial\boldsymbol{\mu}]-\Gamma_2\boldsymbol{\mu}_\perp+\mathbf{k}\Gamma_1(\mu-\mu_\parallel). \qquad (12)$$

It is assumed here, as usual, that the change $|\Delta\mathbf{H}_n(t)|$ of the field at the nucleus is small compared to the equilibrium value of H_n; $\boldsymbol{\mu}_\parallel$ and $\mu_\perp$ are the longitudinal and transverse components of $\boldsymbol{\mu}$ in a coordinate frame tied to the equlibrium direction of $\mathbf{H}_n$; $\mathbf{k}$ is a unit vector along this direction; Γ_1 and Γ_2 are the reciprocal times of the longitudinal and transverse nuclear relaxations.

Determining $\mathbf{M}$ from the first equation of (3) and substituting it in (12) we obtain the nonlinear equations of motion of the nuclear magnetization, which are too complicated to write out here. It follows from the resultant equations that if there is no anisotropy ($H_k=0$) in the region in which solution 2 is realized, the NMR frequency ω_n is zero (because the system energy is not changed by rotation about the z axis). If $H_k\neq 0$, the NMR frequency differs from zero inside this region and vanishes on its boundary.

Let us consider in greater detail the features of excitation and detection of nuclear precession at $H_\Delta=0$. If $T>T_c$, there is no nuclear magnetization and no NMR signal can be observed. At $T<T_c$ we have

$$\omega_n=\gamma A\mu[\eta(\eta-1)]^{1/2},\quad \eta=AM/(AM-H_k). \qquad (13)$$

It can thus be seen that the NMR frequency is proportional to $\mu(T)$. Let the sample be acted upon by a linearly polarized sinusoidal field $\mathbf{h}\sim\exp(i\omega t)$. Solving the equations of motion, we obtain expressions that describe the induced oscillations of $\boldsymbol{\mu}$:

$$\Delta\mu_z=\alpha\gamma A\mu(\eta-1),\quad \Delta\mu_y=\alpha(i\omega+\Gamma_2),\quad \Delta\mu_x=0,$$
$$\alpha=\gamma\mu(\varkappa h_x+h_z)/(\omega_n^2-\omega^2+2i\Gamma_2\omega), \qquad (14)$$
$$\varkappa=\theta\eta=A^2M\mu/(AM-H_k)^2.$$

To simplify the discussion, we use the relation

$$\varkappa h_x+h_z=\lambda\mathbf{hn}, \qquad (15)$$

where $\mathbf{n}$ is a unit vector with components

$$n_x=\varkappa/\lambda,\quad n_y=0,\quad n_z=1/\lambda,\quad \lambda=(1+\varkappa^2)^{1/2}. \qquad (16)$$

Clearly, the nuclear-precession amplitude is maximized when $\mathbf{h}$ is polarized along $\mathbf{n}$. If $\mathbf{h}$ is perpendicular to $\mathbf{n}$ the direct action of $\mathbf{h}$ on the nuclear system is offset by the action of the alternating hyperfine field—A $\mathbf{M}$ and no NMR signal is excited.

Since $\theta\ll 1$ and $\eta>1$, it follows that $\varkappa$ can be larger than or smaller than unity. The vector $\mathbf{n}$ is approximately directed along the x axis if $\varkappa\gg 1$, and along the z axis in the opposite case $\varkappa\ll 1$. The physical reason is the following. The parameter $\varkappa$ acts as the gain of resonant field. The small factor θ enters in the expression for $\varkappa$ because the angle between $\mathbf{M}$ and $\boldsymbol{\mu}$ is equal to $\pi/2-\theta$ in equilibrium. The indirect excitation of the nuclear system via oscillations of the electron magnetization is thus attenuated by a factor $1/\theta$. Nonetheless, the indirect excitation is more effective than the direct one if $\varkappa\gg 1$. For optimal action on the nuclear system the field $\mathbf{h}$ must be so polarized that the amplitude of the $\mathbf{M}$ oscillations be a maximum, i.e., polarized approximately along the x axis. If $\varkappa\ll 1$, direct NMR excitation is more effective than indirect. The field $\mathbf{h}$ must therefore be polarized perpendicular to the equilibrium direction of the nuclear magnetization, i.e., along the z axis.

We write down now the response $\Delta\mathbf{M}$ of the electron magnetic system to the action of the precessing nuclear magnetization at $H_\Delta=0$:

$$\Delta M_x=\varkappa\Delta\mu_z,\quad \Delta M_y=-\Delta\mu_y,\quad \Delta M_z=0. \qquad (17)$$

The total NMR signal is

$$\mathbf{m}=\Delta\boldsymbol{\mu}+\Delta\mathbf{M}. \qquad (18)$$

Since the yth components of $\Delta\mathbf{M}$ and $\Delta\boldsymbol{\mu}$ cancel each other, $\mathbf{m}$ has only two components:

$$m_x=\varkappa\Delta\mu_z,\quad m_z=\Delta\mu_z. \qquad (19)$$

We see therefore that the vector $\mathbf{m}$ is linearly polarized along the $\mathbf{n}$ axis. If $\varkappa\gg 1$, the main contribution to the NMR signal is made by oscillations induced in the electron magnetization by the nuclear system; $\mathbf{m}$ is therefore polarized approximately along the x axis. In the opposite case, $\varkappa\ll 1$, the main contribution to $\mathbf{m}$ is made by the $\boldsymbol{\mu}$ oscillations, so that the vector $\mathbf{m}$ is collinear with the z axis.

CONCLUSION

Let us summarize briefly our results and discuss possible consequences and generalizations.

1. The calculations show that if the internal magnetic fields at the nuclei of a ferromagnet are cancelled out, nuclear magnetic ordering can be observed, accompanied by the onset of a noncollinear electron-nuclear magnetic structure and by substantial changes in the frequencies, the conditions of NMR excitation and detectability. The temperature at which spontaneous nuclear magnetization is produced can be raised considerably by choosing a sample in which the hyperfine field is close in strength to the magnetic-anisotropy field.

The physical mechanism producing this rise in the ordering temperature is the increase of the susceptibility of the subsystem (in this case—the electron magnetization) through which the interaction is effected: this increase is due to the approach of the subsystem to its instability point (to the point of the orientational phase transition of the electron magnetization). It is clear that a similar mechanism can be realized also in other similar situations (for example, it may be capable of raising the superconductivity-transition temperature near a structural phase transition in a crystal).

2. Naturally, all the effects considered here should be observed also for an electron magnetic system describable by two sublattices, ferromagnetic and paramagnetic, if the exchange interaction between the sublattices is antiferromagnetic. Compensation of the exchange field should cause magnetic ordering of the paramagnetic-sublattice spins, owing to the indirect interaction between the spins via the ferromagnetic sublattice. The investigated specimen will then behave as a two-sublattice ferromagnet in the region in which a solution of type (5) is realized, and as a ferromagnet outside this region. Clearly, other conditions being equal, the critical temperature is much higher here than for the nuclear system, since the nuclear gyromagnetic ratio is replaced in (6) by the electron ratio.

3. Cancellation of the internal magnetic fields at the nuclei of a ferromagnet can possibly be used to search for hypothetical arion interactions—a new type of spin-spin interaction, resulting from exchange of massless Higgs bosons (arions).[6] Experiments aimed at observing arion fields are now being performed using NMR in a gas[7] contained in ferromagnetic screen placed in turn in a magnetic field of a solenoid external to the screen. The main idea of these experiments is that the magnetic field produced at the nuclei by the electron spins of the ferromagnetic screen is offset by the magnetic field of the solenoid current; the arion interaction between the electron and nuclear spins, on the other hand, is not cancelled, for in contrast to magnetic fields, no arion fields are produced by currents. After cancelling out as much of the magnetic field as possible, one measures the ratio of the NMR frequencies of two isotopes, a ratio that can be altered by arion interactions.

Cancellation of the hyperfine field at the nuclei of a ferromagnet by a current-induced external field produces a similar situation, viz., the arion field is not cancelled. It is consequently possible in principle to organize experiments aimed at finding NMR arion interactions under conditions when the hyperfine field is cancelled. Such experiments are superior to those in a ferromagnetic screen[7] because the magnetic fields are stronger, and hence the arion fields are also. Naturally, this procedure meets with difficulties of its own. Thus, for example, hyperfine fields at nuclei of different isotopes may turn out to be unequal, and to eliminate this effect it becomes necessary to investigate NMR from diamagnetic impurity atoms, at whose nuclei a magnetic field is produced by magnetic moments of surrounding atoms, and so forth.

The effects that appear when internal fields in a substance are cancelled out can thus be of interest in various branches of physics.

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